

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

**EP 0 765 106 B1**

(12)

**EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention  
of the grant of the patent:  
**27.11.2002 Bulletin 2002/48**

(51) Int Cl.7: **H05B 33/14, C09K 11/06,  
C07C 211/61**

(21) Application number: **96305586.8**

(22) Date of filing: **30.07.1996**

(54) **Light-emitting material for organic electroluminescence device, and organic electroluminescence device for which the light-emitting material is adapted**

Leuchtemittierender Stoff für organische Elektrolumineszenzvorrichtung, und organische Elektrolumineszenzvorrichtung mit diesem leuchtemittierendem dafür geeignetem Stoff

Substance émettant de la lumière pour dispositif organique électroluminescent, et dispositif organique électroluminescent pour lequel la substance est adaptée

(84) Designated Contracting States:  
**DE FR GB**

(30) Priority: **25.09.1995 JP 24560795**  
**29.01.1996 JP 1243096**

(43) Date of publication of application:  
**26.03.1997 Bulletin 1997/13**

(60) Divisional application:  
**01113795.7 / 1 146 034**

(73) Proprietor: **TOYO INK MANUFACTURING CO., LTD.**  
**Tokyo (JP)**

(72) Inventors:  
• **Enokida, Toshio**  
**Chuo-ku, Tokyo (JP)**  
• **Tamano, Michiko**  
**Chuo-ku, Tokyo (JP)**

• **Okutsu, Satoshi**  
**Chuo-ku, Tokyo (JP)**

(74) Representative: **Woods, Geoffrey Corlett et al**  
**J.A. KEMP & CO.**  
**Gray's Inn**  
**14 South Square**  
**London WC1R 5JJ (GB)**

(56) References cited:  
**EP-A- 0 650 955** **EP-A- 0 681 019**  
**EP-A- 0 757 088**

• **DATABASE WPI Section Ch, Week 9618 Derwent Publications Ltd., London, GB; Class E14, AN 96-175691 XP002030009 & JP 08 053 397 A (TOYO INK MFG CO LTD) , 27 February 1996**  
• **POLYMERIC MATERIALS SCIENCE AND ENGINEERING, vol. 72, 1995, US, pages 331-332, XP000671148 L.S.SAPOCHAK & AL: "Luminescent properties of pentacoordinated gallium..."**

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European Patent Convention).

**EP 0 765 106 B1**

## Description

[0001] The present invention relates to a light-emitting material for an organic electroluminescence device (to be sometimes referred to as "EL device" hereinafter) used as a flat light source or a flat display device, and an EL device having a high brightness.

[0002] An EL device using an organic substance is overwhelmingly expected to be usable as a solid light-emitting inexpensive large-screen, full-color display device, and devices are being developed in many ways. Generally, an EL device is composed of a light-emitting layer and a pair of mutually opposite electrodes sandwiching the light-emitting layer. Light emission is the following phenomenon. When an electric field is applied to these two electrodes, the cathode injects electrons into the light-emitting layer and the anode injects holes into the light-emitting layer. When the electrons recombine with the holes in the light-emitting layer, their energy level shifts to a valence bond band to release energy as fluorescent light.

[0003] As compared with inorganic EL devices, conventional organic EL devices require high voltage, and their light emission brightness and light emission efficiency are low. Further, conventional organic EL devices deteriorate in properties to a great extent, and no organic EL device has been put to practical use.

[0004] There has been recently proposed an organic EL device which is produced by laminating a thin film containing an organic compound having a fluorescent quantum effect of emitting light at a low voltage as low as less than 10 V, and it attracts attention (Appl. Phys. Lett., Vol. 51, page 913, 1987).

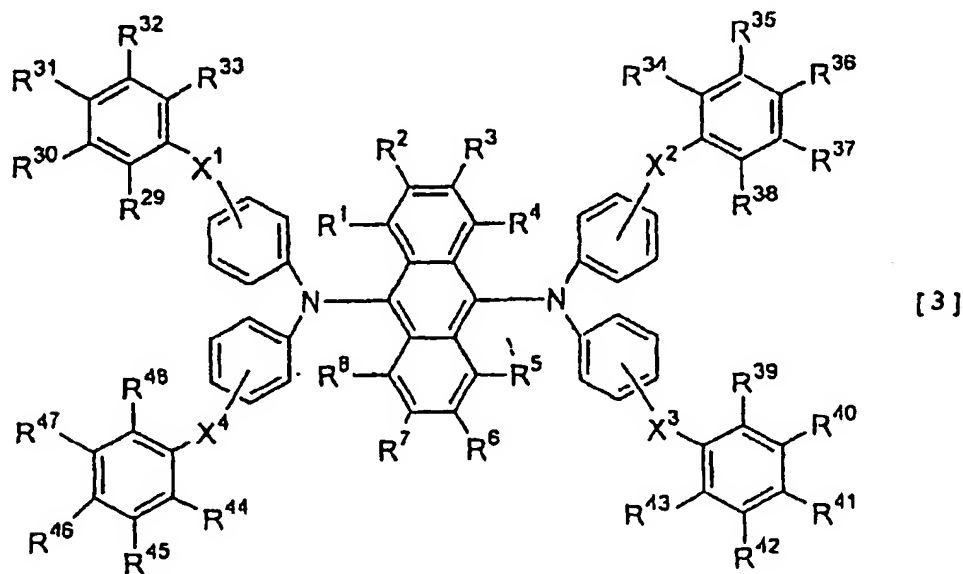
[0005] The above organic EL device has a light-emitting layer containing a metal chelate complex and a hole-injecting layer containing an amine-based compound, and emits green light having a high brightness. The above organic EL device achieves nearly practically usable performance, since it accomplishes a brightness of thousands cd/m<sup>2</sup> and a maximum light emission efficiency of 1.5 lm/W when a direct current voltage of 6 or 7V is applied.

[0006] However, conventional organic EL devices including the above organic EL device are not yet sufficient in brightness improved in brightness to some extent, and their big problem is that they are insufficient in light emission stability in their continuous operation for a long period of time. It is because, for example, the metal chelate complex such as tris(8-hydroxyquinolate)aluminum complex is chemically unstable when light is emitted by the application of an electric field and because the metal chelate complex is poor in adhesion to a cathode that the organic EL device immensely deteriorates in the light-emitting operation for a short time. For developing an organic EL device which has a higher light emission brightness and high light emission efficiency and which has excellent stability in the continuous operation for a long period time, therefore, it is desired to develop a light-emitting material having the excellent light-emitting capability and durability.

[0007] EP-A-650955 relates to particular aromatic amine compounds for use as hole-transporting materials.

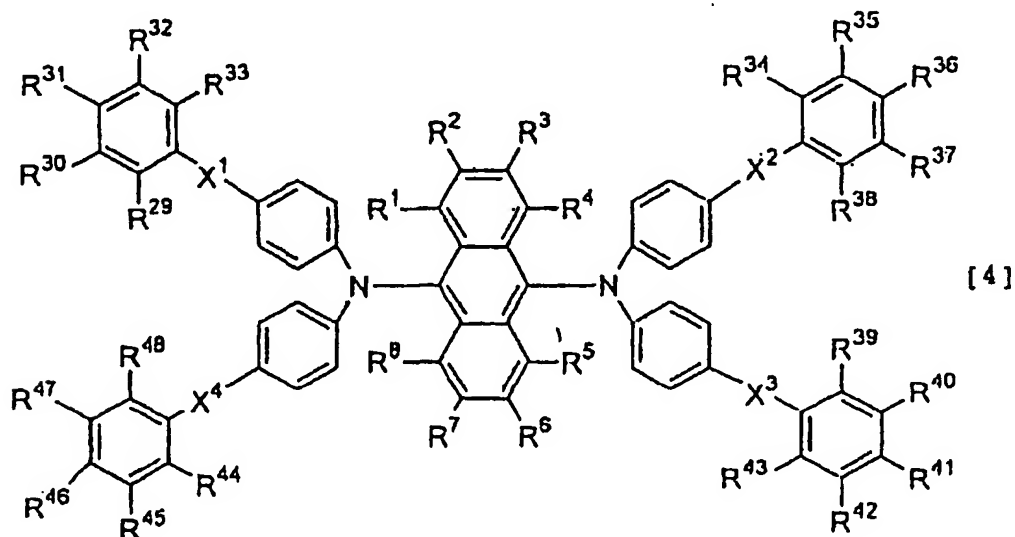
[0008] It is an object of the present invention to provide an organic EL device which is excellent in light emission brightness and light emission effect and which has excellent stability in the repeated operation for a long period of time.

[0009] According to the present invention there is provided a light-emitting compound of general formula [3]



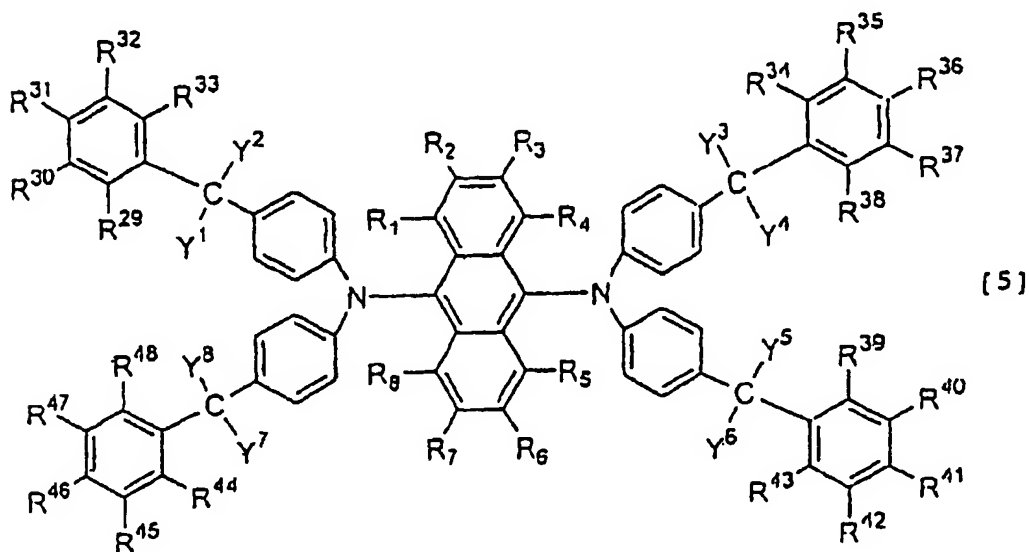
wherein  $R^1$  to  $R^8$  and  $R^{29}$  to  $R^{48}$  are the same or different and each is a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted aryl group or a substituted or unsubstituted amino group, and  $X^1$  to  $X^4$  are the same or different and each is O, S, C=O,  $SO_2$ ,  $(CH_2)_x$ ,  $-O-(CH_2)_y$ , a substituted or unsubstituted alkylene group or a substituted or unsubstituted alicyclic residue, provided that  $x$  and  $y$  are not both 0, and wherein adjacent substituents selected from  $R^1$  to  $R^4$  or  $R^5$  to  $R^8$  optionally form an aryl ring together.

[0010] Typically, the compound of formula [3] is a compound of formula [4],



wherein  $R^1$  to  $R^8$ ,  $R^{29}$  to  $R^{48}$  and  $X^1$  and  $X^4$  are as defined above.

[0011] According to the present invention, further, there is provided a light-emitting compound of general formula [5]



wherein each of  $R^1$  to  $R^8$  and  $R^{29}$  to  $R^{48}$  are as defined above and each of  $Y^1$  to  $Y^8$ , which may be the same or different, is a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted aryl group having 6 to 16 carbon atoms.

[0012] Also provided is the use of the compounds of the invention as light-emitting materials in an organic electro-

luminescence device.

[0013] The light-emitting compounds of the present invention may be used in an organic electroluminescence device comprising a light-emitting layer or a plurality of thin organic compound layers including the light-emitting layer between a pair of electrodes which are an anode and a cathode, wherein the light-emitting layer comprises, as light-emitting material, a compound of the invention.

[0014] Preferably the above organic electroluminescence device comprises an organic compound layer between the light-emitting layer and the anode, which organic compound layer comprises an aromatic tertiary amine derivative or a phthalocyanine derivative.

[0015] It is preferred that the aromatic tertiary amine derivative has the following general formula [6],



wherein each of  $B^1$  to  $B^4$ , which may be the same or different, is a substituted or unsubstituted aryl group having 6 to 16 carbon atoms, and Z is a substituted or unsubstituted arylene group.

[0016] It is also preferable that the above organic electroluminescence device comprises an organic compound layer between the light-emitting layer and the cathode, which organic compound layer comprises a metal complex compound or a nitrogen-containing five-membered derivative.

[0017] Preferably the metal complex compound has the following general formula [7],



wherein each of  $Q^1$  and  $Q^2$ , which may be the same or different, is a substituted or unsubstituted hydroxyquinoline derivative or a substituted or unsubstituted hydroxybenzoquinoline derivative, and L is a ligand which is halogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted aryl or heteroaryl group containing a nitrogen atom, -OR in which R is a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group or a substituted or unsubstituted aryl or heteroaryl group containing a nitrogen atom, or L is -O-Ga- $Q^3(Q^4)$  in which  $Q^3$  and  $Q^4$ , which may be the same or different, are as defined for  $Q^1$  and  $Q^2$ .

[0018] The light-emitting compounds of the present invention are preferably used in an organic electroluminescence device obtained by forming a plurality of thin organic compound layers including a light-emitting layer between a pair of electrodes, wherein the light-emitting layer contains a compound of the invention, the device has an organic layer containing a compound of the general formula [6] between the light-emitting layer and the anode, and the device has an organic layer containing a compound of the general formula [7] between the light-emitting layer and the cathode.

[0019] In each of the general formulae [3] to [5], carbon atom(s) of the aryl group may be replaced with at least one of a nitrogen atom, an oxygen atom and a sulfur atom.

[0020] In the compounds of the general formulae [3] to [5], each of  $R^1$  to  $R^{48}$  is independently a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted aryl group or a substituted or unsubstituted amino group. In each of  $R^1$  to  $R^{48}$ , carbon atom(s) of the aryl group may be replaced with at least one of a nitrogen atom, an oxygen atom and a sulfur atom.

[0021] Specific examples of  $R^1$  to  $R^{48}$  are as follows. The halogen atom includes fluorine, chlorine, bromine and iodine. The substituted or unsubstituted alkyl group includes unsubstituted alkyl groups having 1 to 20 carbon atoms

such as methyl, ethyl, propyl, butyl, sec-butyl, tert-butyl, pentyl, hexyl, heptyl, octyl and stearyl, and substituted alkyl groups having 1 to 20 carbon atoms such as 2-phenylisopropyl, trichloromethyl, trifluoromethyl, benzyl,  $\alpha$ -phenoxybenzyl,  $\alpha,\alpha$ -dimethylbenzyl,  $\alpha,\alpha$ -methylphenylbenzyl,  $\alpha,\alpha$ -ditrifluoromethylbenzyl, triphenylmethyl and  $\alpha$ -benzyloxybenzyl. The substituted or unsubstituted alkoxy group includes unsubstituted alkoxy groups having 1 to 20 carbon atoms such as methoxy, ethoxy, propoxy, n-butoxy, t-butoxy, n-octyloxy and t-octyloxy, and substituted alkoxy groups having 1 to 20 carbon atoms such as 1,1,1-tetrafluoroethoxy, phenoxy, benzyloxy and octylphenoxy. The substituted or unsubstituted aryl group includes substituted or unsubstituted aryl groups having 6 to 18 carbon atoms such as phenyl, 2-methylphenyl, 3-methylphenyl, 4-methylphenyl, 4-ethylphenyl, biphenyl, 4-methylbiphenyl, 4-ethylbiphenyl, 4-cyclohexylbiphenyl, terphenyl, 3,5-dichlorophenyl, naphthyl, 5-methylnaphthyl, anthryl, pyrenyl, and aryl groups whose carbon atom is replaced with a nitrogen atom, etc., such as furanyl, thiophenyl, pyrrolyl, pyranlyl, thiopyranlyl, pyridinyl, thiazolyl, imidazolyl, pyrimidinyl, triazinyl, indolyl, quinolyl, purinyl and carbazolyl. The substituted or unsubstituted amino group includes amino, dialkylamino groups such as dimethylamino and diethylamino, phenylmethylamino, diphenylamino, ditolylamino and dibenzylamino. Further, adjacent substituents ( $R^1$  to  $R^{48}$ ) may form phenyl, naphthyl, anthryl or pyrenyl.

[0022] In the compounds of the general formulae [3] and [4], each of  $X^1$  to  $X^4$  is independently O, S, C=O,  $SO_2$ ,  $(CH_2)_x-O-(CH_2)_y$ , a substituted or unsubstituted alkylene group or a substituted or unsubstituted alicyclic residue, provided that each of x and y is independently an integer of 0 to 20 and that  $x + y = 0$  in no case. Substituents of the substituted alkylene group or the substituted alicyclic residue are those which are specified as  $R^1$  to  $R^{48}$ . The alkylene group of the substituted or unsubstituted alkylene group includes alkylene groups having 1 to 20 carbon atoms. The substituted alkylene group preferably includes 2-phenylisopropylene, dichloromethylene, difluoromethylene, benzylene,  $\alpha$ -phenoxybenzylene,  $\alpha,\alpha$ -dimethylbenzylene,  $\alpha,\alpha$ -methylphenylbenzylene, diphenylmethylene and  $\alpha$ -benzyloxybenzylene. The substituted or unsubstituted alicyclic residue includes divalent alicyclic residues having 5 to 7 carbon atoms such as cyclopentyl, cyclohexyl, 4-methylcyclohexyl and cycloheptyl.

[0023] In the compound of the general formula [5], each of  $Y^1$  to  $Y^8$  is independently a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted aryl group having 6 to 16 carbon atoms. Specific examples of the substituted or unsubstituted alkyl group and the substituted or unsubstituted aryl group are those described concerning the above  $R^1$  to  $R^{48}$ .

[0024] In the above compounds of the general formulae [3] to [5], the compounds which have substituents each of which has an aromatic ring, or the compounds in which adjacent substituents indicated by  $R^1$  to  $R^{48}$  form aromatic rings, have increased glass transition points and melting points, and these compounds therefore show improved resistance (heat resistance) against Joule's heat generated in the organic layers, between the organic layers, or between the organic layer and the electrode of a metal. When the above compounds are used as a light-emitting material for an organic EL device, they exhibit a high light emission brightness and are advantageous in the continuous light-emitting operation for a long period of time. Examples of the above compounds of the general formulae [3] to [5] in which adjacent substituents indicated by  $R^1$  to  $R^{48}$  form aromatic rings are those in which  $R^1$  and  $R^2$  are mutually fused,  $R^3$  and  $R^4$  are mutually fused,  $R^5$  and  $R^6$  are mutually fused and  $R^7$  and  $R^8$  are mutually used, to form benzene rings, naphthalene rings, anthracene rings or pyrene rings. The compounds of the present invention shall not be limited to the above substituents.

[0025] The compounds of the general formulae [3] to [5] are synthesized, for example, by the following method.

[0026] 9,10-Dihalogenoanthracene, an amine derivative which may have a substituted and potassium carbonate are allowed to react in a solvent in the presence of a catalyst, whereby the compounds of the general formulae [3] to [5] are synthesized. The anthracene derivative may be replaced with an anthraquinone derivative. The potassium carbonate may be replaced with sodium carbonate, potassium hydroxide, sodium hydroxide or aqueous ammonia. The catalyst is selected from a copper powder, a copper (I) chloride, tin, tin (II) chloride, pyridine, aluminum chloride or titanium tetrachloride. The solvent is selected from benzene, toluene or xylene. The method of synthesizing the compounds of the general formula [3] to [5] shall not be limited to the above method.

[0027] Specific examples of the compounds of the general formula [3] to [5] are shown in Table 1, while the compounds of the general formula [3] to [5] shall not be limited thereto.

Table 1

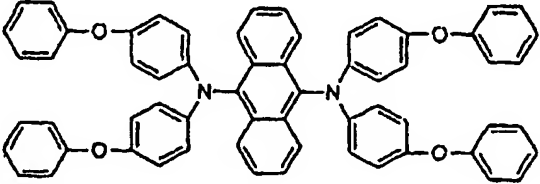
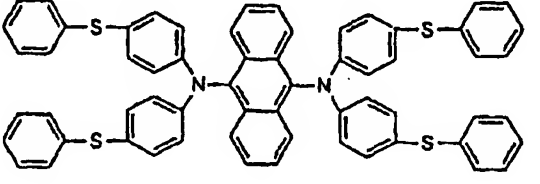
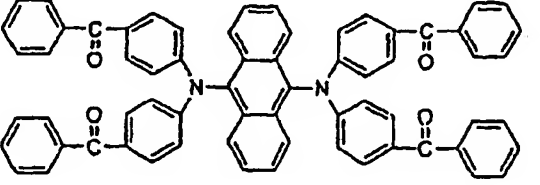
(14)	
(15)	
(16)	

Table 1 (continued)

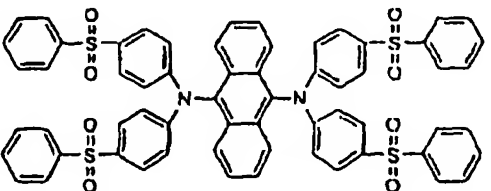
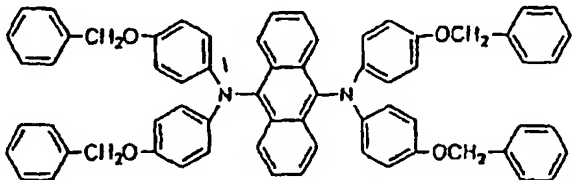
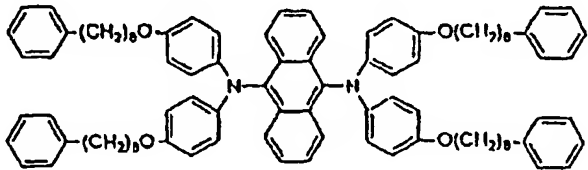
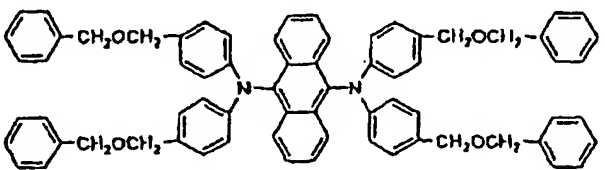
Compound	Chemical structure
(17)	
(18)	
(19)	
(20)	

Table 1 (continued)

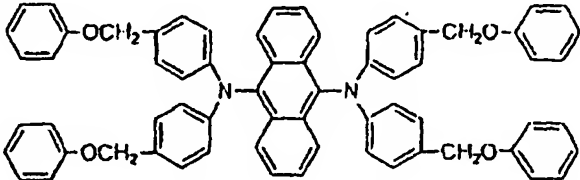
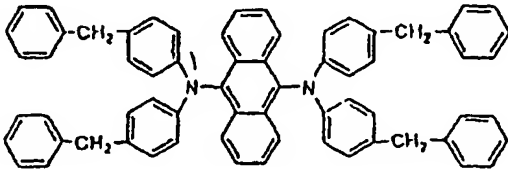
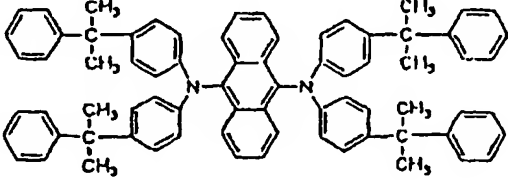
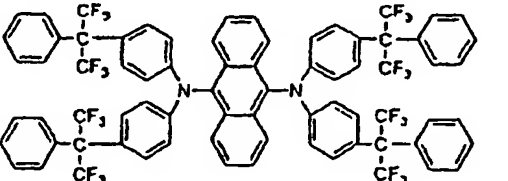
Compound	Chemical structure
(21)	
(22)	
(23)	
(24)	



Table 1 (continued)

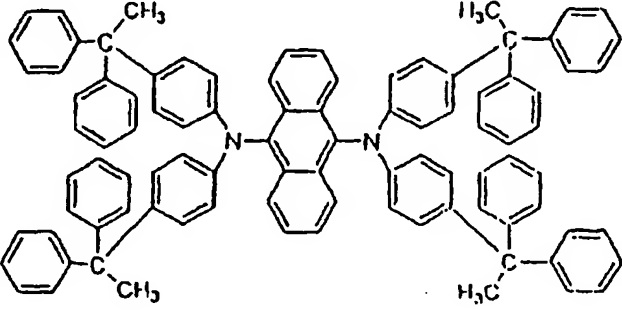
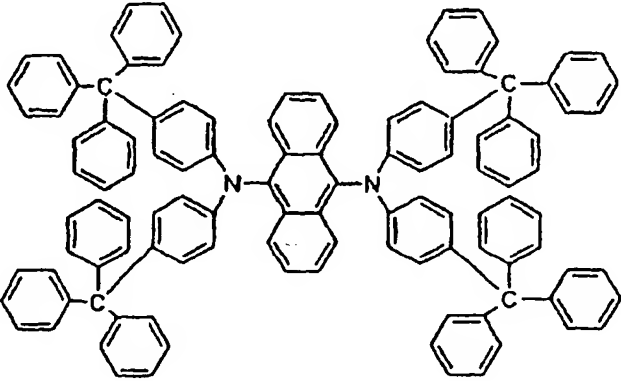
Compound	Chemical structure
(25)	
(26)	

Table 1 (continued)

Compound	Chemical structure
(27)	
(28)	
(31)	
(32)	

[0028] The compounds of the general formula [3] to [5], provided by the present invention, have intense fluorescence and are excellent in light emission when an electric field is applied. Further, the compounds of the general formula [3] to [5] can be effectively used as light-emitting materials since they have the excellent capability of being injected with, and transporting, holes from an electrode of a metal, and have the excellent capability of being injected with, and transporting, electrons from an electrode of a metal. Moreover, each of the compounds of the general formula [3] to [5] may be used in combination with other hole-transporting material, other electron-transporting material or other dopant.

[0029] The organic EL device is classified into an organic EL device of one layer type which has one organic thin layer between an anode and a cathode and an organic EL device of a multi-layer type which has a plurality of organic thin layers between an anode and a cathode. When the organic EL device is a mono-layer type, it has a light-emitting layer between an anode and a cathode. The light-emitting layer contains a light-emitting material. For transporting holes injected from the anode to the light-emitting material or transporting electrons injected from the cathode to the light-emitting material, the light-emitting layer may further contain a hole-injecting material or an electron-injecting material. However, each of the light-emitting materials of the present invention alone can be used for forming a light-emitting layer, since they have all of a very high light-emitting quantum efficiency, a very high hole-injecting and hole-transporting capability and a high electron-injecting and electron-transporting capability, and are formable into a thin layer. For example, the organic EL device of a multi-layer type has a layer structure such as anode/hole-injecting layer/light-emitting layer/cathode, anode/light-emitting layer/electron-injecting layer/cathode, or anode/hole-injecting layer/

light-emitting layer/electron-injecting layer/cathode. Each of the compounds of the general formula [3] to [5] can be used as a light-emitting material in the light-emitting layer since they have high light emission properties and the properties of injecting and transporting holes and injecting and transporting electrons.

[0030] The light-emitting layer may contain a known light-emitting material, a known dopant, a known hole-injecting material and a known electron-injecting material as required in combination with any one of the compounds of the general formula [3] to [5]. A multi-layer structure of the organic EL device can serve to prevent quenching from degrading the brightness and the device life. A light-emitting material, a dopant, a hole-injecting material and an electron-injecting material may be used in combination as required. Further, some dopants improve the brightness and efficiency in light emission, and serve to obtain light emission in red or blue. Further, each of the hole-injecting layer, the light-emitting layer and electron-injecting layer may have a structure of at least two layers. For example, when the hole-injecting layer has a structure of two layers, a layer into which holes are injected from an electrode will be referred to as "hole-injecting layer", and a layer which receives holes from the "hole-injecting layer" and transports the holes to the light-emitting layer will be referred to as "hole-transporting layer". Similarly, when the electron-injecting layer has a structure of two layers, a layer into which electrons are injected from an electrode will be referred to as "electron-injecting layer", and a layer which receives electrons from the "electron-injecting layer" and transports the electrons to the light-emitting layer will be referred to as "electron-transporting layer". Materials for these layers are selected depending upon factors such as their energy level, heat resistance and adhesion to other organic layer or an electrode of a metal.

[0031] The light-emitting material or the doping material used in combination with the compounds of the general formula [3] to [5] includes anthracene, naphthalene, phenanthrene, pyrene, tetracene, coronene, fluorescein, perylene, phthaloperylene, naphthaloperylene, perinone, naphthaloperinone, diphenylbutadiene, tetraphenylbutadiene, coumarin, oxadiazole, aldazine, bisbenzoxazoline, bisstyryl, pyrazine, cyclopentadiene, quinoline metal complex, aminoquinoline metal complex, benzoquinoline metal complex, imine, diphenylethylene, vinyl anthracene, diaminocarbazole, pyrane, thiopyrane, polymethine, merocyanine, an imidazole-chelated oxynoid compound, quinacridone, rubrene and fluorescence dyes, although the above materials shall not be limited to these.

[0032] The doping material has a high fluorescence intensity when it is in the state of a low concentration, and it has a decreased fluorescence intensity when it is in the state of a high concentration, particularly, in the state of a solid. Light emission is therefore effectively achieved by incorporating a low concentration of the doping material into a fluorescence material as a host, and it can be therefore expected to increase the light emission efficiency. Further, some doping materials can be used for shifting the light color from the fluorescence color of a host material to the fluorescence color of a doping material on the basis of the energy shift from the host material.

[0033] Although differing depending upon a host material used, the amount of the doping material is 0.0001 to 50 % by weight, preferably 0.001 to 5 % by weight.

[0034] The hole-injecting material is selected from compounds which are capable of transporting holes, receiving holes from an anode, injecting the holes into the light-emitting layer or a light-emitting material, preventing the movement of excitons generated in the light-emitting layer into the electron-injecting layer or the electron-injecting material and forming a thin film. Specifically, the above hole-injecting material includes a phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, oxazole, oxadiazole, triazole, imidazole, imidazolone, imidazolthione, pyrazoline, pyrazolone, tetrahydroimidazole, hydrazone, acylhydrazone, polyarylethane, stilbene, butadiene, benzidine type triphenylamine, styrylamine type triphenylamine, diamine type triphenylamine, derivatives of these, and polymer materials such as polyvinylcarbazole, polysilane and an electrically conductive polymer, although the above hole-injecting material shall not be limited to these.

[0035] In the hole-injecting material that can be used for the organic EL device of the present invention, more effective are aromatic tertiary amine derivatives or phthalocyanine derivatives. Specific examples thereof include triphenylamine, tritolylamine, tolyldiphenylamine, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N,N',N'-(4-methylphenyl)-1,1'-phenyl-4,4'-diamine, N,N,N',N'-(4-methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N'-diphenyl-N,N'-dinaphthyl-1,1'-biphenyl-4,4'-diamine, N,N'-(methylphenyl)-N,N'-(4-n-butylphenyl)-phenanthrene-9,10-diamine, N,N-bis(4-di-4-tolylaminophenyl)-4-phenylcyclohexane, and oligomers or polymers having an aromatic tertiary amine skeleton thereof.

[0036] In the compound of the general formula [6] which can be used as hole-injecting material in the present invention, B<sup>1</sup> to B<sup>4</sup> is independently a substituted or unsubstituted aryl group having 6 to 16 carbon atoms. Specific examples of B<sup>1</sup> to B<sup>4</sup> include aromatic ring groups such as phenyl, biphenyl, terphenyl, naphthyl, anthryl, phenanthryl, fluorenyl and pyrenyl, and these aromatic ring groups may contain a substituent. Carbon atom(s) of the above aryl group may be replaced with at least one of N, O and S atoms. In the aryl group, a combination of B<sup>1</sup> and B<sup>2</sup> or a combination of B<sup>3</sup> and B<sup>4</sup> may form a ring including N in the general formula [6]. Examples of the aryl groups whose carbon atom is replaced with a nitrogen atom, etc., include furanyl, thiophenyl, pyrrolyl, pyranal, thiopyranal, pyridyl, thiazolyl, imidazolyl, pyrimidinyl, triazinyl, indolyl, quinolyl, purinyl and carbazolyl. Z is a divalent arylene group. Examples of the arylene group includes divalent aromatic ring groups such as phenylene, biphenylene, terphenylene, naphthylene, anthrylene, phenanthrylene, fluorenylene and pyrenylene groups. The above arylene groups may have the same sub-

stituents as R<sup>1</sup> to R<sup>48</sup> in arbitrary sites. Typical examples of the general formula [6] and other compounds which are more effective hole-injecting materials are shown in Table 2, while the present invention shall not be limited to these examples.

Table 2

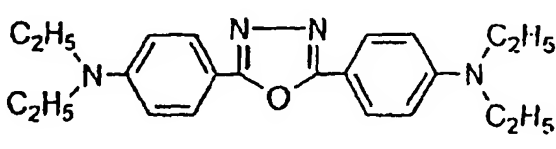
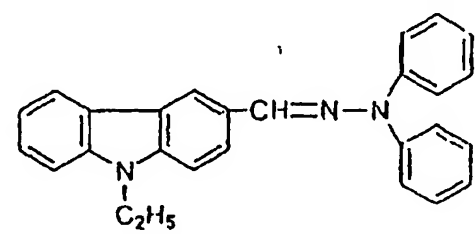
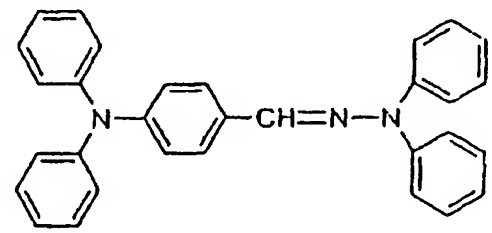
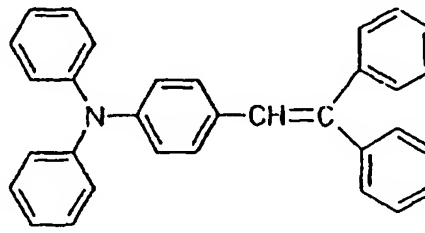
Compound	Chemical structure
A - 1	
A - 2	
A - 3	
A - 4	

Table 2 (continued)

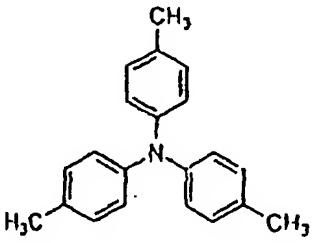
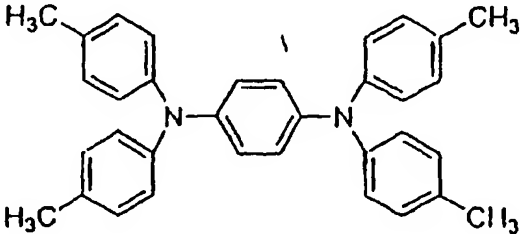
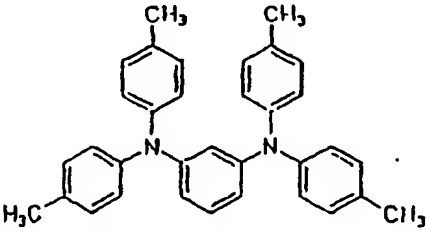
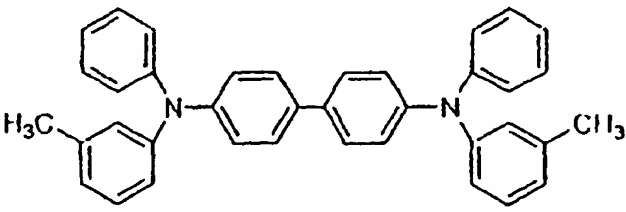
Compound	Chemical structure
A - 5	 <chem>Cc1ccc(N(c2ccc(C)cc2)c3ccc(C)cc3)c4ccc(C)cc4</chem>
A - 6	 <chem>Cc1ccc(N(c2ccc(C)cc2)c3ccc(N(c4ccc(C)cc4)c5ccc(C)cc5))cc3</chem>
A - 7	 <chem>Cc1ccc(N(c2ccc(C)cc2)c3ccc(N(c4ccc(C)cc4)c5ccc(C)cc5))cc3</chem>
A - 8	 <chem>Cc1ccc(N(c2ccccc2)c3ccccc3N4c5ccccc5c6ccccc64)cc1</chem>

Table 2 (continued)

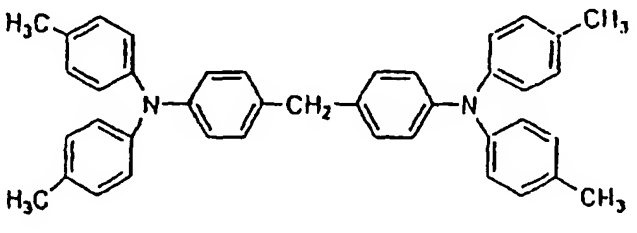
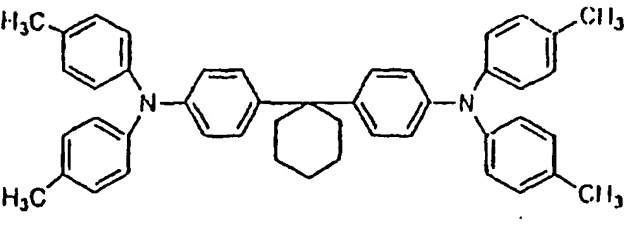
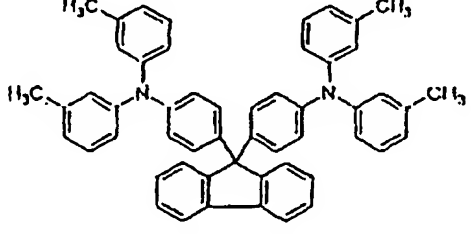
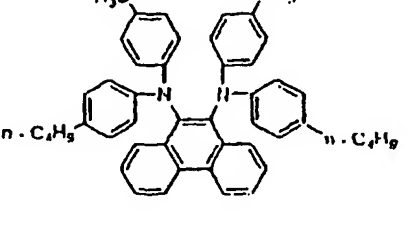
Compound	Chemical structure
Λ - 9	
Λ - 10	
Λ - 11	
Λ - 12	

Table 2 (continued)

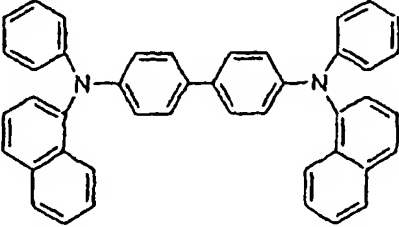
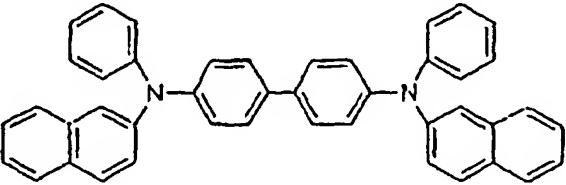
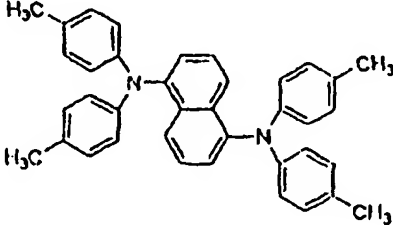
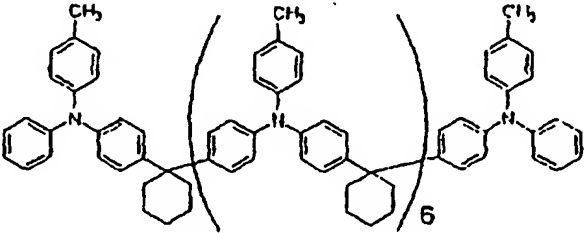
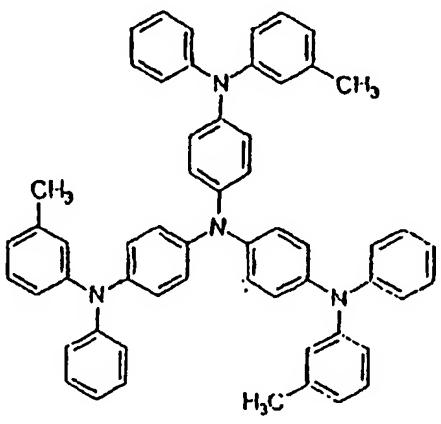
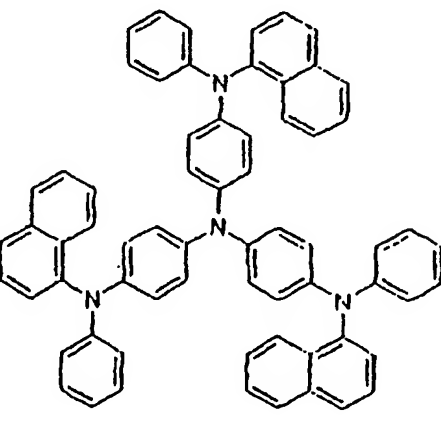
Compound	Chemical structure
$\Lambda - 13$	
$\Lambda - 14$	
$\Lambda - 15$	
$\Lambda - 16$	

Table 2 (continued)

Compound	Chemical structure
$\Lambda - 17$	
$\Lambda - 18$	

[0037] Examples of the phthalocyanine (Pc) derivative include  $H_2Pc$ ,  $CuPc$ ,  $CoPc$ ,  $NiPc$ ,  $ZnPc$ ,  $PdPc$ ,  $FePc$ ,  $MnPc$ ,  $ClAlPc$ ,  $ClGaPc$ ,  $ClInPc$ ,  $ClSnPc$ ,  $Cl_2SiPc$ ,  $(HO)AlPc$ ,  $(HO)CaPc$ ,  $VOpc$ ,  $TiOPc$ ,  $MoOPc$ ,  $GaPc-O-CaPc$  and naphthalocyanine derivatives in which the phthalocyanine skeletons of the above phthalocyanine derivatives are replaced with naphthalocyanine skeletons, while the phthalocyanine derivative shall not be limited to these.

[0038] The electron-injecting material is selected from compounds which are capable of transporting electrons, receiving electrons from a cathode, injecting the electrons into the light-emitting layer or a light-emitting material, preventing the movement of excitons generated in the light-emitting layer into the hole-injecting layer or the hole-injecting material and forming a thin film. For example, the above electron-injecting material includes fluorenone, anthraquinodimethane, diphenylquinone, thiopyran dioxide, oxazole, oxadiazole, triazole, imidazole, perylenetetracarboxylic acid, fluorenylidene methane, anthraquinodimethane, anthrone and derivatives of these, while the electron-injecting material shall not be limited to these. Further, the hole-transporting material may be sensitivity-increased by incorporating an electron-accepting material, and the electron-injecting material may be sensitivity-increased by incorporating an elec-



tron-donating material.

[0039] In the organic EL device of the present invention, the following metal complex compounds or nitrogen-containing five-membered derivatives are more effective electron-injecting materials. Specific examples of the metal complex compounds include lithium 8-hydroxyquinolate, zinc bis(8-hydroxyquinolate), copper bis(8-hydroxyquinolate), manganese bis(8-hydroxyquinolate), aluminum tris(8-hydroxyquinolate), aluminum tris(2-methyl-(8-hydroxyquinolate), gallium tris(8-hydroxyquinolate), beryllium bis(10-hydroxybenzo[h]quinolate), zinc bis(10-hydroxybenzo[h]quinolate), chlorogallium bis(2-methyl-8-quinolate), gallium bis(2-methyl-8-quinolate)(o-cresolate), aluminum bis(2-methyl-8-quinolate)(1-naphtholate), and gallium bis(2-methyl-8-quinolate)(2-naphtholate), while the metal complex compounds shall not be limited to these. The nitrogen-containing five-membered derivatives are preferably oxazole, thiazole, oxadiazole, thiadiazole and triazole derivatives. Specific examples thereof include 2,5-bis(1-phenyl)-1,3,4-oxazole, dimethyl POPOP, 2,5-bis(1-phenyl)-1,3,4-thiazole, 2,5-bis(1-phenyl)-1,3,4-oxadiazole, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-oxadiazole, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole, 1,4-bis[2-(5-phenyloxadiazolyl)]benzene, 1,4-bis[2-(5-phenyloxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-2,3,4-thiadiazole, 2,5-bis(1-naphthyl)-1,3,4-thiadiazole, 1,4-bis[2-(5-phenylthiadiazolyl)]benzene, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-triazole, 2,5-bis(1-naphthyl)-1,3,4-triazole, and 1,4-bis[2-(5-phenyltriazolyl)]benzene, while the above derivatives shall not be limited to these.

[0040] In the organic EL device of the present invention, the compound of the general formula [7] is a more effective electron-injecting material. In the general formula [7], each of Q<sup>1</sup> and Q<sup>2</sup> is independently a hydroxyquinone derivative such as 8-hydroxyquinoline, 8-hydroxyquinoline, 8-hydroxy-2-phenylquinoline, 8-hydroxy-5-methylquinoline, 8-hydroxy-3,5,7-trifluoroquinoline. L is a halogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group, a substituted or unsubstituted aryl group which may contain a nitrogen atom, -OR in which R is a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted cycloalkyl group or a substituted or unsubstituted aryl group which may contain a nitrogen atom, or -O-Ga-Q<sup>3</sup>(Q<sup>4</sup>) in which Q<sup>3</sup> and Q<sup>4</sup> have the same meanings as those of Q<sup>1</sup> and Q<sup>2</sup>. The above halogen atom, alkyl group, cycloalkyl group, aryl group whose ring may contain a nitrogen atom, and the alkyl group, cycloalkyl group and aryl group whose ring may contain a nitrogen atom are those described concerning the general formulae [1] to [5].

[0041] Typical examples of the compound of the general formula [7] and typical examples of the electron-injecting material used in the present invention will be shown in the following Table 3, while the above compound and the above material shall not be limited to these.

Table 3

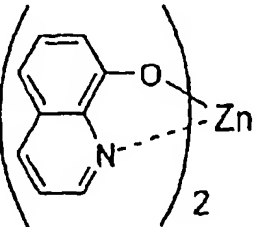
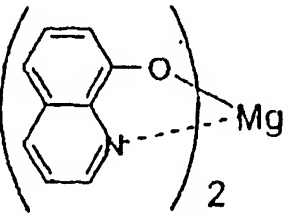
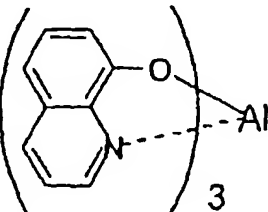
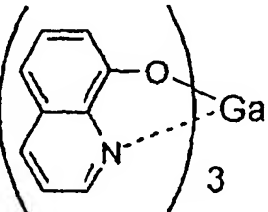
Compound	Chemical structure
B-1	
B-2	
B-3	
B-4	

Table 3 (continued)

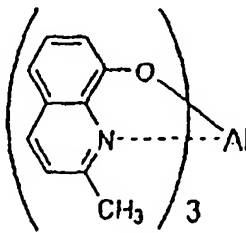
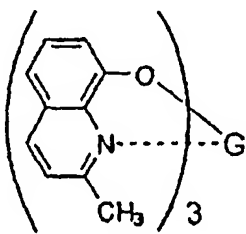
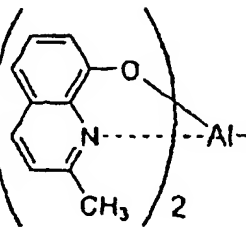
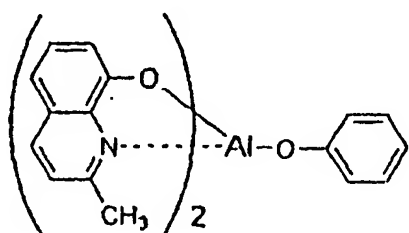
Compound	Chemical structure
B-5	
B-6	
B-7	
B-8	

Table 3 (continued)

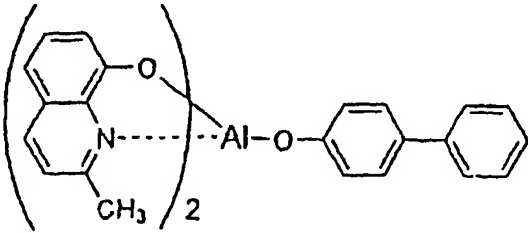
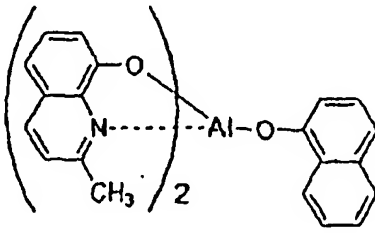
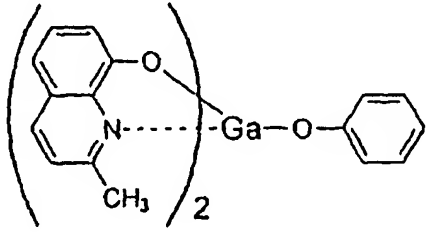
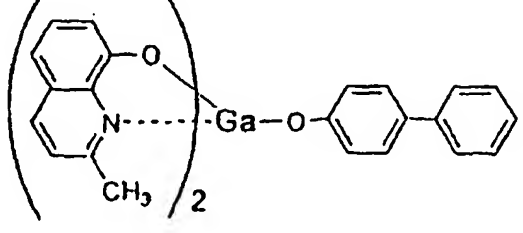
Compound	Chemical structure
B-9	
B-10	
B-11	
B-12	

Table 3 (continued)

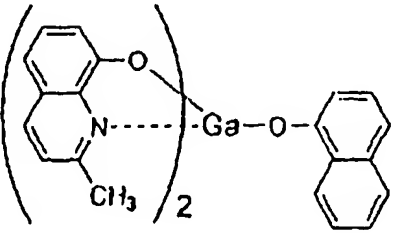
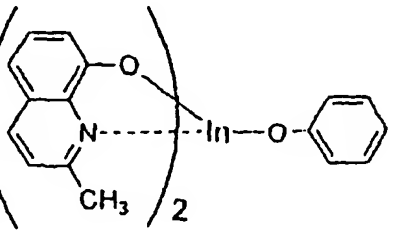
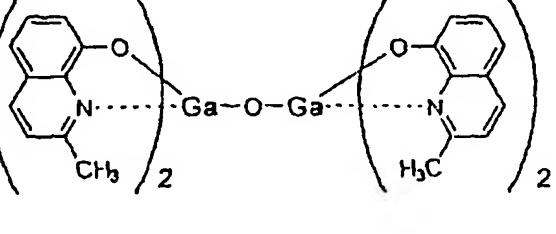
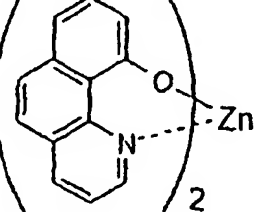
Compound	Chemical structure
B-13	
B-14	
B-15	
B-16	

Table 3 (continued)

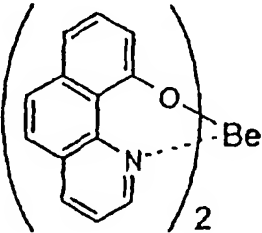
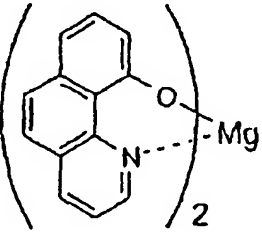
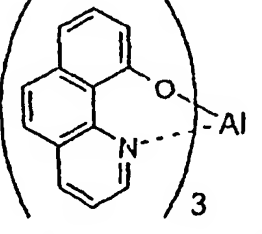
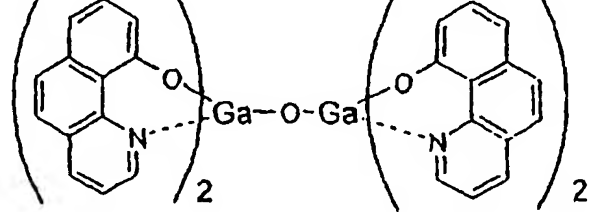
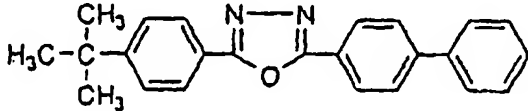
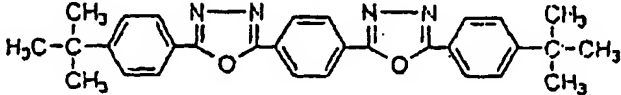
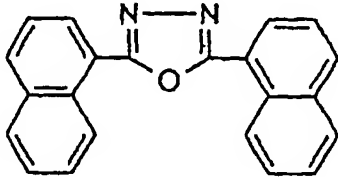
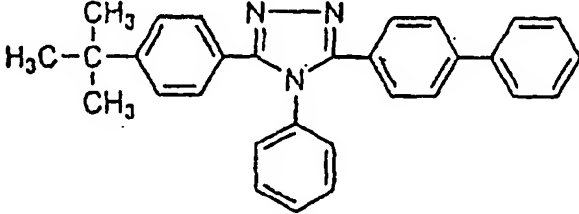
Compound	Chemical structure
B-17	
B-18	
B-19	
B-20	

Table 3 (continued)

Compound	Chemical structure
B - 2 1	
B - 2 2	
B - 2 3	
B - 2 4	

[0042] In the organic EL device of the present invention, the light-emitting layer may contain at least one of other light-emitting material, dopant, hole-injecting or hole-transporting material and electron-injecting or electron-transporting material in combination with any one of the compounds of the general formulae [3] to [5]. For improving the organic EL device of the present invention in the stability against temperature, humidity and ambient atmosphere, a protective layer may be formed on the surface of the device, or the device as a whole may be sealed with a silicone oil, a resin, or the like.

[0043] The electrically conductive material used for the anode of the organic EL device is preferably selected from those materials having a work function of greater than 4 eV. This electrically conductive material includes carbon, aluminum, vanadium, iron, cobalt, nickel, tungsten, silver, gold, platinum, palladium, alloys of these, metal oxides such as tin oxide and indium oxide used for ITO substrates or NESA substrates, and organic electrically conductive resins such as polythiophene and polypyrrole.

[0044] The electrically conductive material used for the cathode is preferably selected from those having a work function of smaller than 4 eV. This electrically conductive material includes magnesium, calcium, tin, lead, titanium,

yttrium, lithium, ruthenium, manganese, aluminum and alloys of these, while it shall not be limited to these. Typical examples of the alloys include magnesium/silver, magnesium/indium and lithium/aluminum, while the alloys shall not be limited to these. The metal ratio of the alloy is properly selected depending upon the temperature of a deposition source, atmosphere and the degree of vacuum. Each of the anode and the cathode may have a structure of at least two layers as required.

[0045] For the effective light emission of the organic EL device, at least one of the electrodes is desirably transparent in the light emission wavelength region of the device. Further, the substrate is desirably transparent. The transparent electrode is produced from the above electrically conductive material by a deposition method or a sputtering method such that a predetermined transparency is secured. The electrode which forms a light emission surface preferably has a light transmittance of at least 10 %. The substrate is not specially limited if it has adequate mechanical and thermal strength and is transparent. For example, it is selected from transparent resin substrates such as a glass substrate, a polyethylene substrate, a polyethylene terephthalate substrate, a polyether sulfone substrate and a polypropylene substrate.

[0046] Each of the layers forming the organic EL device of the present invention can be formed by any one of dry film forming methods such as a vacuum deposition method, a sputtering method, a plasma method and an ion plating method and wet film forming methods such as a spin coating method, a dipping method and a flow coating method. The thickness of each layer is not specially limited, while each layer is required to have a proper thickness. When the layer thickness is too large, inefficiently, a high voltage is required to achieve predetermined emission of light. When the layer thickness is too small, the layer is liable to have a pinhole, etc., so that sufficient light emission brightness is hard to obtain when an electric field is applied. Generally, the thickness of each layer is preferably in the range of from 5 nm to 10  $\mu$ m, more preferably 10 nm to 0.2  $\mu$ m.

[0047] In the wet film forming method, a material for forming an intended layer is dissolved or dispersed in a proper solvent, and a thin film is formed from the solution or dispersion. The solvent is selected from chloroform, tetrahydrofuran and dioxane, while the solvent shall not be limited to these. For improving the film formability and preventing the occurrence of pinholes, the above solution or dispersion for forming the layer may contain a proper resin and a proper additive. The resin suitable for use in the present invention includes insulating resins such as polystyrene, polycarbonate, polyarylate, polyester, polyamide, polyurethane, polysulfone, polymethyl methacrylate, polymethyl acrylate and cellulose, copolymers of these, photoconductive resins such as poly-N-vinylcarbazole and polysilane, and electrically conductive resins such as polythiophene and polypyrrole. The above additive includes an antioxidant, an ultraviolet absorbent and a plasticizer.

[0048] When the light-emitting layer of the organic EL device is formed of the compound of the present invention, and when organic EL device has a specific hole-injecting layer or electron-injecting layer in combination, the organic EL device is improved in properties such as light emission efficiency, maximum light emission brightness. Further, the organic EL device is highly stable against heat and electric current, and further it gives practically usable light emission brightness at a low actuation voltage, so that the deterioration of the device, a vital problem of prior art devices, can be remarkably decreased.

[0049] The organic EL device of the present invention can be applied to a flat panel display of a wall-mountable TV set, a flat light-emitter, a light source for a copying machine or a printer, a light source for a liquid crystal display or a counter, a display board and a sign lamp. The organic EL device of the present invention is therefore greatly industrially valuable.

[0050] Further, the compounds of the present invention can be also used in the fields of an organic EL device, an electrophotographic photoreceptor, a photoelectric converter, a solar cell and an image sensor.

[0051] The present invention will be explained with reference to Examples hereinafter, in which "%" stands for "% by weight" and "part" stands for "part by weight".

#### Method of Synthesis of Compound (23)

[0052] 15 Parts of 9,10-dilidoanthracene, 27 parts of 4,4-diisopropyl(2-phenyl)diphenylamine, 12 parts of potassium carbonate and 0.8 parts of a copper powder were placed in a flask, and the mixture was stirred under heat at 200°C for 30 hours. Then, the reaction mixture was diluted with 500 parts of water, and then the mixture was extracted with chloroform. A chloroform layer was concentrated, purified by silica gel column chromatography and re-precipitated in n-hexane to give 18 parts of a powder having yellow fluorescence. The powder was analyzed for a molecular weight to show that the powder was Compound (23). The results of elemental analysis of the product were as shown below. Results of elemental analysis

As  $C_{74}H_{68}N_2$

Calculated (%)	C: 90.24,	H: 6.91,	N: 2.85
----------------	-----------	----------	---------



(continued)

Found (%)	C: 90.59,	H: 6.81,	N: 2.60
-----------	-----------	----------	---------

## 5 Comparative Examples 1 to 5 and Examples 1 and 2

[0053] Compound (A-16) shown in Table 2 was vacuum-deposited on a cleaned glass plate with an ITO electrode to form a hole-injecting layer having a thickness of 20 nm. Then, a compound shown in the following Table 4 as a light-emitting material was vacuum-deposited to form a light-emitting layer having a thickness of 20 nm. The chemical structures of compounds 1 to 4 and 13 used in Comparative Examples 1 to 5 are shown in the following Table 1a. Further, gallium bis (2-methyl-8-quinolate)(1-phenolate) complex was vacuum-deposited to form an electron-injecting layer having a thickness of 20 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. Each layer was formed by vacuum deposition under a vacuum of  $1.33 \times 10^{-4}$  Pa ( $10^{-6}$  Torr) at a temperature of substrate temperature. Table 4 shows light emission characteristics of the so-obtained EL devices. The EL devices were measured for brightness at a direct current voltage of 5 V, and all of the EL devices showed a maximum brightness of at least 10,000 cd/m<sup>2</sup>. Those compounds of the general formula [3] in which R<sup>1</sup> to R<sup>8</sup> were aryl groups or adjacent substituents formed aromatic rings had high glass transition points and high melting points and gave EL devices which showed excellent initial brightness and device lives when driven to emit light.

Table 1a

Compound	Chemical structure
(1)	
(2)	
(3)	
(4)	

Table 1a (continued)

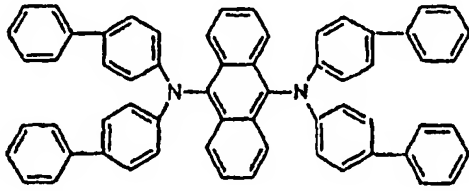
Compound	Chemical structure
(13)	

Table 4

Example	Compound	Light emission brightness (cd/m <sup>2</sup> )	Light emission efficiency (lm/W)
Comp. Example 1	1	450	1.20
Comp. Example 2	2	430	1.15
Comp. Example 3	3	470	1.11
Comp. Example 4	4	420	1.05
1	23	750	1.85
Comp. Example 5	13	700	1.55
2	14	750	1.75

## Example 3

[0054] Compound (A-16) shown in Table 2 was vacuum-deposited on a cleaned glass plate with an ITO electrode to form a hole-injecting layer having a thickness of 20 nm, and Compound (23) as a light-emitting material, shown in Table 1, was vacuum-deposited thereon to form a light-emitting layer having a thickness of 20 nm. Then, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole was vacuum-deposited to form an electron-injecting layer having a thickness of 20 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The organic EL device showed a green light emission of 770 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 27,000 cd/m<sup>2</sup> and a light emission efficiency of 1.80 (lm/W).

## Example 4

[0055] An organic EL device was obtained in the same manner as in Example 3 except that a 5 nm thick hole-injecting layer of metal-free phthalocyanine was formed between the ITO electrode and the compound (A-16). The organic EL device showed a green light emission of 1,200 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 29,000 cd/m<sup>2</sup> and a light emission efficiency of 1.70 (lm/W). When compared with the organic EL device obtained in Example 3, the organic EL device obtained in Example 4 showed a high brightness on a low voltage side.

## Example 5

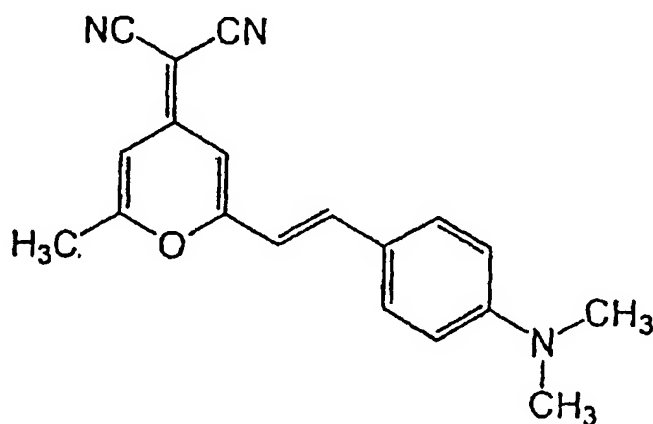
[0056] An organic EL device was obtained in the same manner as in Example 3 except that the hole-injecting layer of Compound (A-16) was replaced with a 20 nm thick hole-injecting layer of metal-free phthalocyanine. The organic EL device showed a green light emission of 650 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 15,000 cd/m<sup>2</sup> and a light emission efficiency of 1.30 (lm/W).

## Example 6

[0057] An organic EL device was obtained in the same manner as in Example 1 except that the 20 nm thick light-emitting layer of Compound (23) was replaced with a 10 nm thick light-emitting layer formed by vapor-depositing Compound (23) and Compound (C-4) shown in Table 6 in a Compound (23)/Compound (C-4) weight ratio of 100/1. The organic EL device showed a green light emission of 700 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 35,000 cd/m<sup>2</sup> and a light emission efficiency of 2.70 (lm/W).

## Example 7

[0058] An organic EL device was obtained in the same manner as in Example 1 except that the 20 nm thick light-emitting layer of Compound (23) was replaced with a 10 nm thick light-emitting layer formed by vapor-depositing Compound (23) and the following compound in a Compound (23)/the following compound weight ratio of 100/1. The organic EL device showed an orange light emission of 500 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 18,000 cd/m<sup>2</sup> and a light emission efficiency of 1.65 (lm/W).



## Comparative Examples 6 and 7 and Examples 8 to 30

[0059] A hole-injecting material was vacuum-deposited on a cleaned glass plate with an ITO electrode under conditions shown in Table 5 to form a hole-injecting layer having a thickness of 30 nm. Then, a light-emitting material was vacuum-deposited to form a light-emitting layer having a thickness of 30 nm. Further, an electron-injecting material was vacuum-deposited to form an electron-injecting layer having a thickness of 30 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. Each layer was formed by vacuum deposition under a vacuum of  $1.33 \times 10^{-4}$  Pa ( $10^{-6}$  Torr) at a temperature of substrate temperature. Table 5 shows light emission characteristics of the so-obtained EL devices. The EL devices were measured for brightness at a direct current voltage of 5 V, and all of the EL devices had high-brightness characteristics, as high as a maximum brightness of at least 10,000 cd/m<sup>2</sup>. The compounds of the general formula [3] to [5] gave EL devices which showed excellent initial brightness and device lives when driven to emit light. Further, in the layer structure of the organic EL device, those EL devices which used a combination of any one of the light-emitting materials of the general formulae [3] to [5] with the hole-injecting material of the general formula [6] and the electron-injecting material of the general formula [7] showed the most excellent characteristics.

Ex.	Hole-injecting material	Light-emitting material	Electron-injecting layer	Light emission brightness (cd/m <sup>2</sup> )	Maximum brightness (cd/m <sup>2</sup> )	Maximum light emission efficiency (lm/W)
Comp. Example 6	(A-13)	(1)	(B-11)	1,100	23,000	2.8

(continued)

Ex.	Hole-injecting material	Light-emitting material	Electron-injecting layer	Light emission brightness (cd/m <sup>2</sup> )	Maximum brightness (cd/m <sup>2</sup> )	Maximum light emission efficiency (lm/W)
Comp. Example 7	(A-13)	(13)	(B-11)	2,200	32,000	2.5
8	(A-13)	(14)	(B-11)	3,000	58,000	4.3
9	(A-13)	(18)	(B-11)	3,500	78,000	5.1
10	(A-13)	(22)	(B-11)	3,500	69,000	5.0
11	(A-13)	(23)	(B-11)	5,100	110,000	12
12	(A-13)	(24)	(B-11)	5,000	100,000	10
13	(A-13)	(25)	(B-11)	5,600	125,000	10
14	(A-13)	(26)	(B-11)	4,800	78,000	8.2
15	(A-13)	(32)	(B-11)	3,800	58,000	7.3
16	(A-13)	(23)	(B-1)	2,200	29,000	3.1
17	(A-13)	(23)	(B-4)	1,460	15,000	1.9
18	(A-13)	(23)	(B-5)	1,800	19,000	2.4
19	(A-13)	(23)	(B-6)	1,600	18,500	2.2
20	(A-13)	(23)	(B-10)	2,220	21,000	3.1
21	(A-13)	(23)	(B-12)	4,800	89,000	8.0
22	(A-13)	(23)	(B-13)	4,500	77,000	7.7
23	(A-13)	(23)	(B-15)	3,900	65,300	7.3
24	(A-13)	(23)	(B-16)	3,700	33,000	4.1
25	(A-13)	(23)	(B-21)	2,100	20,000	3.2
26	(A-13)	(23)	(B-22)	2,500	21,000	3.1
27	(A-13)	(23)	(B-23)	2,800	22,000	2.5
28	(A-13)	(23)	(B-24)	2,400	23,000	2.4
29	(A-12)	(23)	(B-11)	5,000	108,000	11
30	(A-8)	(14)	(B-11)	4,500	71,000	6.9
Light emission brightness = brightness at a direct current voltage of 5 (V).						

## 45 Example 31

[0060] A hole-injecting material (A-4) was vacuum-deposited on a cleaned glass plate with an ITO electrode to form a hole-injecting layer having a thickness of 30 nm. Then, a hole-injecting material (A-13) was vacuum-deposited to form a second hole-injecting layer having a thickness of 10 nm. Then, Compound (24) as a light-emitting material was vacuum-deposited to form a light-emitting layer having a thickness of 30 nm. Further, an electron-injecting material (B-12) was vacuum-deposited to form an electron-injecting layer having a thickness of 30 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The organic EL device showed a green light emission of 1,100 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 87,000 cd/m<sup>2</sup> and a light emission efficiency of 9.3 (lm/W).

55

## Example 32

[0061] A hole-injecting material (A-16) was vacuum-deposited on a cleaned glass plate with an ITO electrode to form

a hole-injecting layer having a thickness of 20 nm. Then, Compound (23) as a light-emitting material was vacuum-deposited to form a light-emitting layer having a thickness of 20 nm. Further, Compound (B-23) as an electron-injecting material was vacuum-deposited to form an electron-injecting layer having a thickness of 20 nm. An electrode having a thickness of 150 nm was formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The organic EL device showed a green light emission of 770 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 27,000 cd/m<sup>2</sup> and a light emission efficiency of 1.8 (lm/W).

#### Example 33

[0062] An organic EL device was obtained in the same manner as in Example 31 except that a 5 nm thick hole-injecting layer of metal-free phthalocyanine was formed between the ITO electrode and Compound (A-16). The organic EL device showed a green light emission of 1,200 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 29,000 cd/m<sup>2</sup> and a light emission efficiency of 1.70 (lm/W).

#### Example 34

[0063] An organic EL device was obtained in the same manner as in Example 31 except that the hole-injecting layer of Compound (A-16) was replaced with a 20 nm thick hole-injecting layer of metal-free phthalocyanine. The organic EL device showed a green light emission of 650 cd/m<sup>2</sup> at a direct current voltage of 5 V, a maximum brightness of 15,000 cd/m<sup>2</sup> and a light emission efficiency of 1.30 (lm/W).

#### Examples 35 - 44

[0064] An organic EL device was obtained in the same manner as in Example 31 except that the light-emitting layer was replaced with a 20 nm thick light-emitting layer formed by vapor-depositing Compound (23) and a compound shown in the following Table 6. Table 7 shows light emission characteristics of the so-obtained organic EL devices. The organic EL devices were measured for brightness at a direct current voltage of 5 V, and each organic EL device had high-brightness characteristics, as high as a maximum brightness of at least 10,000 cd/m<sup>2</sup> and emitted an intended color.

Table 6

Compound	Chemical structure
C - 1	
C - 2	
C - 3	
C - 4	
C - 5	

Table 6 (continued)

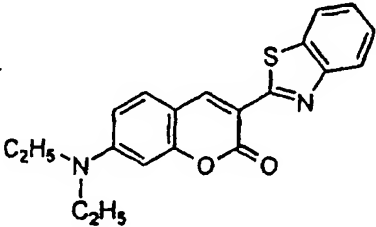
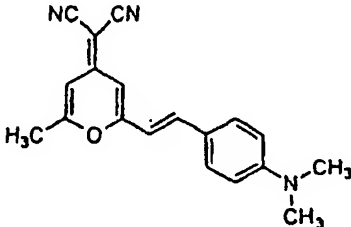
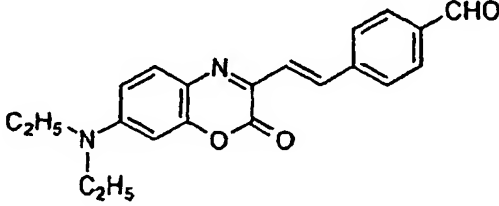
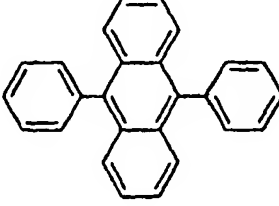
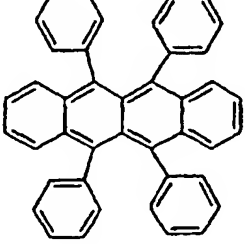
Compound	Chemical structure
C - 6	 <chem>CCN(CC)c1ccc2c(c1)oc(=O)c(c2)c3nc4ccccc4s3</chem>
C - 7	 <chem>CN(C)c1ccc(/C=C/c2cc(OC(=C2C#N)C#N)cc1C</chem>
C - 8	 <chem>CCN(CC)c1ccc2c(c1)oc(=O)c(c2)/C=C/c3ccc(C=O)cc3</chem>
C - 9	 <chem>c1ccc(cc1)-c2cc3ccccc3cc2-c4ccccc4</chem>
C - 10	 <chem>c1ccc(cc1)-c2cc3cc4ccccc4cc3cc2-c5ccccc5</chem>

Table 7

Example	Dopant in Table 6	Light emission brightness (cd/m <sup>2</sup> )	Maximum brightness (cd/m <sup>2</sup> )	Maximum light emission efficiency (lm/W)	Color of emitted light
35	(C-1)	1,000	28,000	2.7	Green
36	(C-2)	850	25,000	2.7	Green
37	(C-3)	900	28,000	2.6	Green
38	(C-4)	700	35,000	2.7	Green
39	(C-5)	600	21,000	3.1	Green
40	(C-6)	500	22,000	3.0	Green
41	(C-7)	1,100	36,000	4.1	Orange
42	(C-8)	1,500	38,000	4.0	Red
43	(C-9)	900	30,000	3.7	Blue
44	(C-10)	1,100	31,000	3.5	Yellow

[0065] The organic EL devices obtained in the above Examples showed a light emission brightness of at least 10,000 cd/m<sup>2</sup> and high light emission efficiency. When the organic EL devices obtained in the above Examples were allowed to continuously emit light at 3 mA/cm<sup>2</sup>, all the organic EL devices emitted light stably for more than 1,000 hours and showed almost no dark spots, while the light emission brightness (above 100 cd/m<sup>2</sup>) of conventional organic EL devices decreased to 1/2 or less of the initial brightness within 500 hours. Further, the comparative organic EL devices showed many dark spots, and the number of dark spots increased, and the sizes thereof increased, with the passage of light emission time. Since the light-emitting materials of the present invention have a remarkably high fluorescence quantum effect, the organic EL devices using these light-emitting materials were able to emit light having high brightness in a range where a low voltage was applied. Further, when the dopant was used in light-emitting layers in combination with the compounds of the general formulae [3] to [5], the organic EL devices were improved in maximum light emission brightness and maximum light emission efficiency. Further, when the dopant for the red or blue light emission was added to the compounds of the general formulae [3] to [5] for the light emission of bluish green, green or yellow, the organic EL devices emitted red or blue light.

[0066] The organic EL device of the present invention is intended to achieve improvements in light emission efficiency and light emission brightness and an increase in device life, and is not to impose any limitation on other light-emitting material, dopant, hole-injecting or hole-transporting material, electron-injecting or electron-transporting material, sensitizer, resin, electrode material, and the like which are used in combination with the compounds of the present invention, nor is it to impose any limitation on the method of the production thereof.

[0067] The organic EL device for which any one of the light-emitting materials of the present invention is applied emits light having a high brightness with high light emission efficiency, and achieves a long device life, as compared with conventional devices. Therefore, the organic EL device which has at least one layer formed of any one of the light-emitting material of the present invention and has a device constitution according to the present invention shows a high brightness, high light emission efficiency and long device life. Throughout this specification, it shall be understood that:

[0068] Alkyl groups may be straight or branched and may in general contain from 1 to 20, preferably to 10, more preferably 1 to 8, for example 1 to 6 carbon atoms. Substituted alkyl groups include haloalkyl, for example chloro- and fluoro-alkyl groups which may comprise one or more halogen substituents. Substituted alkyl groups also include aryl substituted alkyl groups containing one or more aryl substituents. Such groups include aralkyl groups such as benzyl and phenethyl.

[0069] Alkoxy groups may be straight or branched and may in general contain from 1 to 20, preferably to 10, more preferably 1 to 8, for example 1 to 6 carbon atoms. Substituted alkoxy groups include halogen-substituted and aryl-substituted alkoxy groups. Halo-substituted alkoxy includes fluoro- and chloro- substituted alkoxy and may comprise one or more halogen substituents. Aryl-substituted alkoxy may comprise one or more aryl substituents.

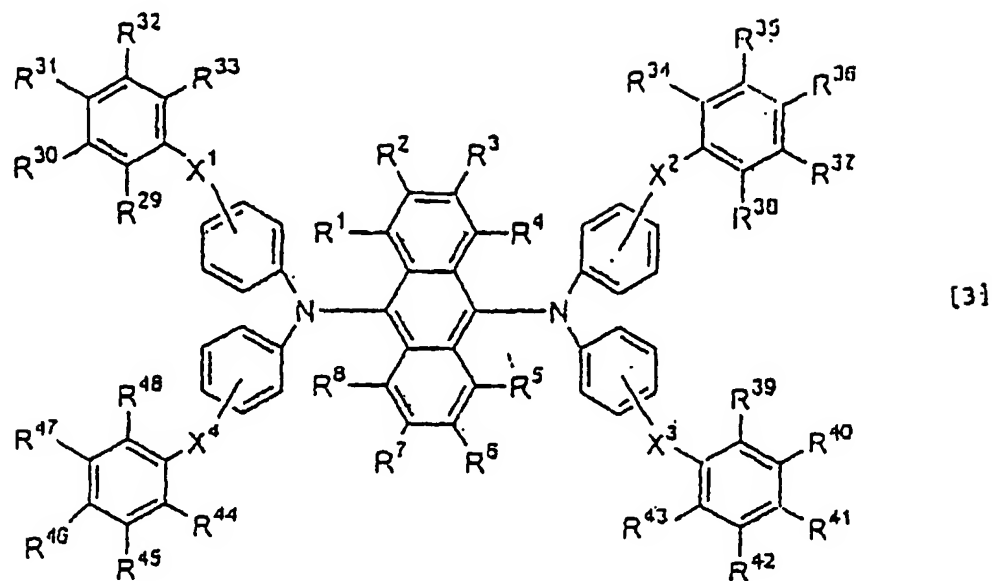
[0070] Aryl groups may be unsubstituted or substituted by one or more substituents, such as an alkyl or haloalkyl group or a halogen atom.

[0071] Amino groups may be unsubstituted or substituted. Substituted amino include secondary alkylamino and arylamino groups and tertiary dialkylamino, alkylarylamino and diarylamino groups.



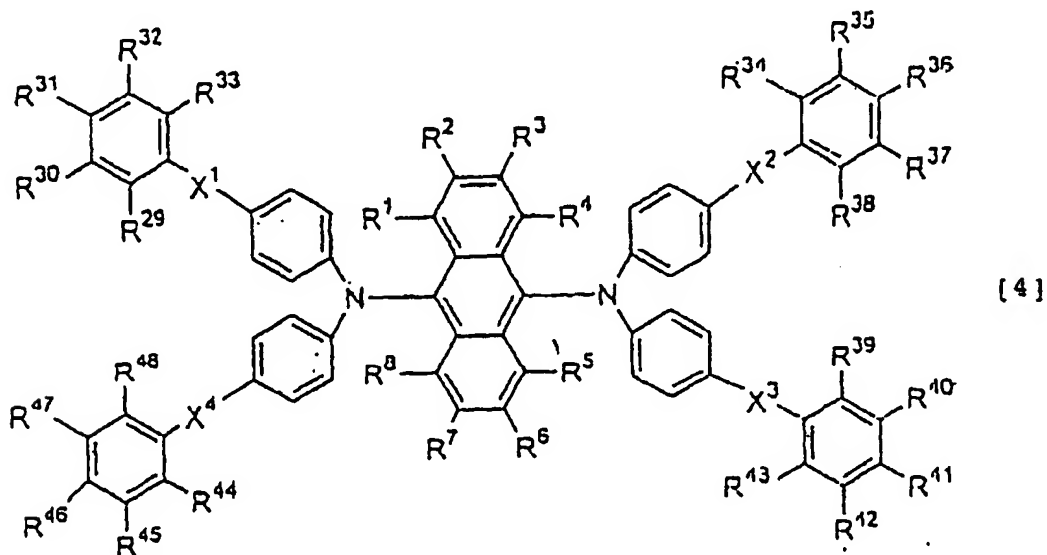
## Claims

1. A light emitting compound of general formula [3]



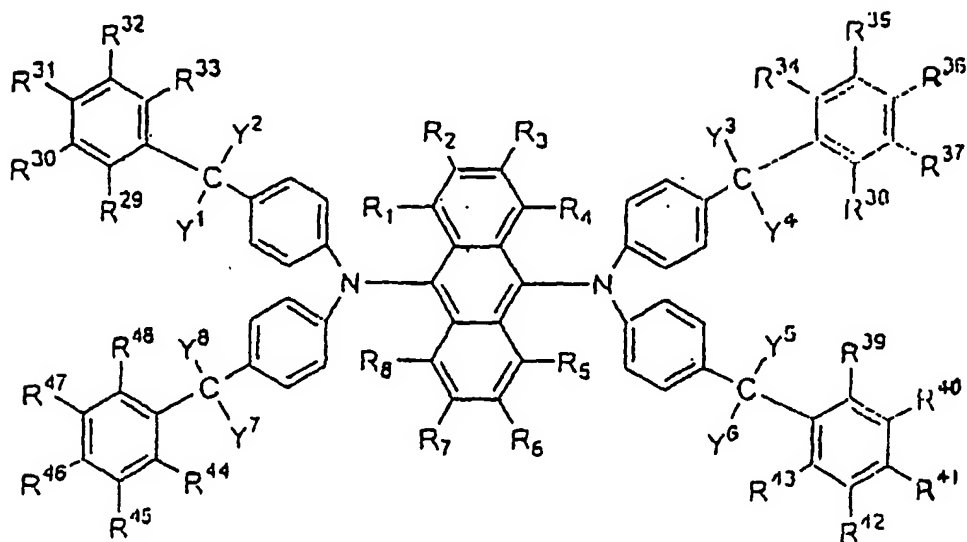
wherein  $R^1$  to  $R^8$  and  $R^{29}$  to  $R^{48}$  are the same or different and each is a hydrogen atom, a halogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted aryl group or a substituted or unsubstituted amino group, and  $X^1$  to  $X^4$  are the same or different and each is O, S, C=O, SO<sub>2</sub>, (CH<sub>2</sub>)<sub>x</sub>-O-(CH<sub>2</sub>)<sub>y</sub>, a substituted or unsubstituted alkylene group or a substituted or unsubstituted alicyclic residue, provided that x and y are not both 0, and wherein adjacent substituents selected from  $R^1$  to  $R^4$  or  $R^5$  to  $R^8$  optionally form an aryl ring together.

2. A compound according to claim 1 of general formula [4]



wherein  $R^1$  to  $R^8$ ,  $R^{29}$  to  $R^{48}$  and  $X^1$  to  $X^4$  are as defined in claim 1.

3. A light-emitting compound of general formula [5]

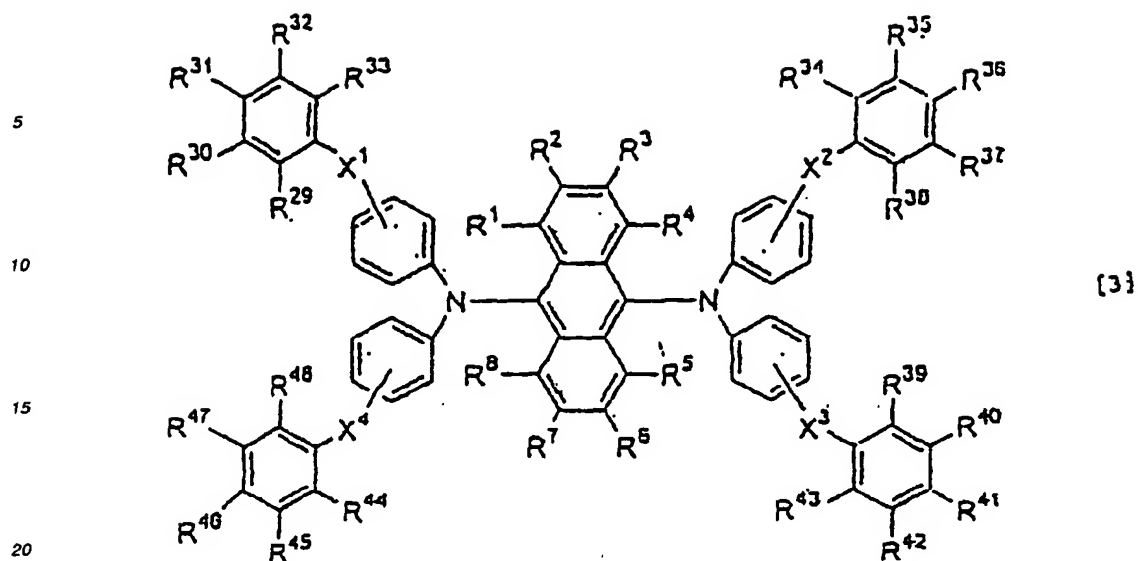


wherein each of  $R^1$  to  $R^8$  and  $R^{29}$  to  $R^{48}$  are as defined in claim 1 and each of  $Y^1$  to  $Y^8$ , which may be the same or different, is a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms or a substituted or unsubstituted aryl group having 6 to 16 carbon atoms.

4. Use of a compound according to any one of claims 1 to 3 as a light-emitting material in an organic electroluminescence device.

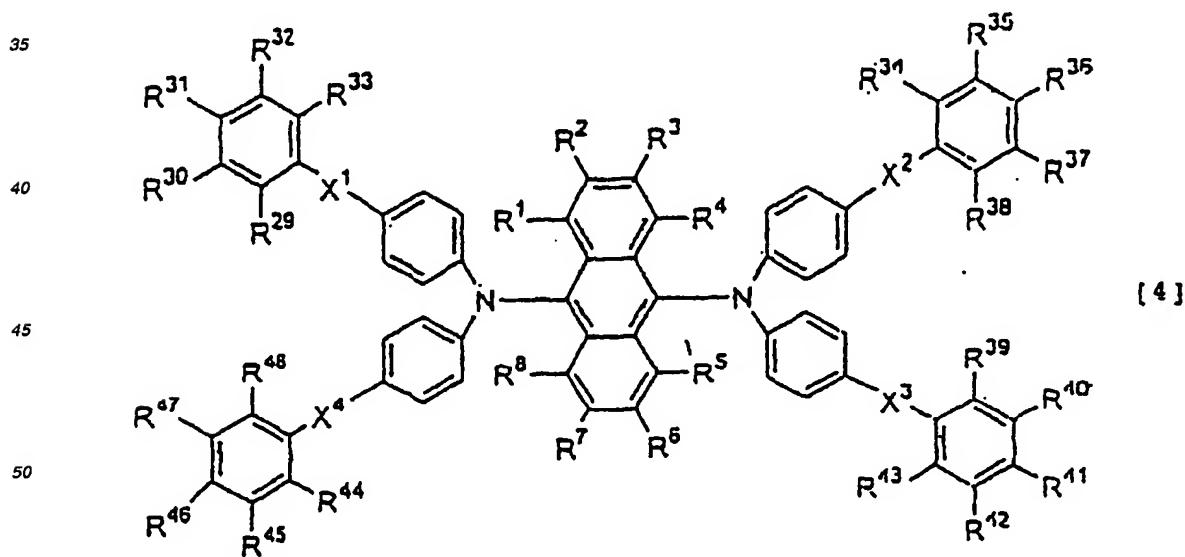
**Patentansprüche**

1. Lichtemittierende Verbindung der allgemeinen Formel [3]



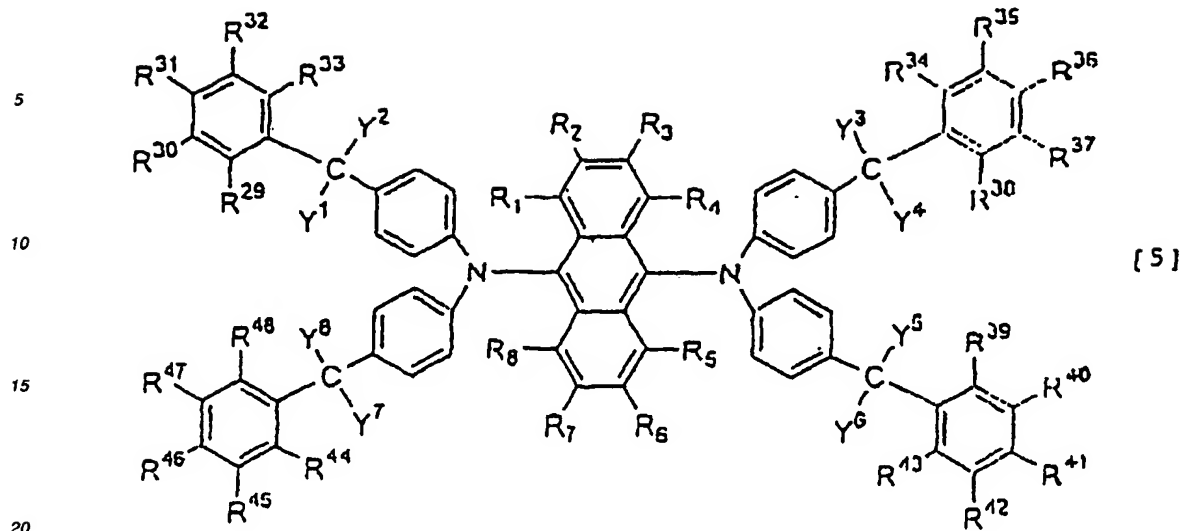
25 worin  $R^1$  bis  $R^8$  und  $R^{29}$  bis  $R^{48}$  gleich oder verschieden sind und jedes ein Wasserstoffatom, ein Halogenatom, eine substituierte oder unsubstituierte Alkylgruppe, eine substituierte oder unsubstituierte Alkoxygruppe, eine substituierte oder unsubstituierte Arylgruppe oder eine substituierte oder unsubstituierte Aminogruppe ist, und  $X^1$  bis  $X^4$  gleich oder verschieden sind und jedes O, S, C=O,  $SO_2$ ,  $(CH_2)_x-O-(CH_2)_y$ , eine substituierte oder unsubstituierte Alkylengruppe oder ein substituiertes oder unsubstituiertes alicyclisches Rest ist, vorausgesetzt, dass X und y nicht beide 0 sind, und worin benachbarte Substituenten, gewählt aus  $R^1$  bis  $R^4$  oder  $R^5$  bis  $R^8$ , wahlweise zusammen einen Arylring bilden.

30 2. Verbindung nach Anspruch 1 der allgemeinen Formel [4]



55 worin  $R^1$  bis  $R^8$ ,  $R^{29}$  bis  $R^{48}$  und  $X^1$  bis  $X^4$  wie in Anspruch 1 definiert sind.

3. Lichtemittierende Verbindung der allgemeinen Formel [5]

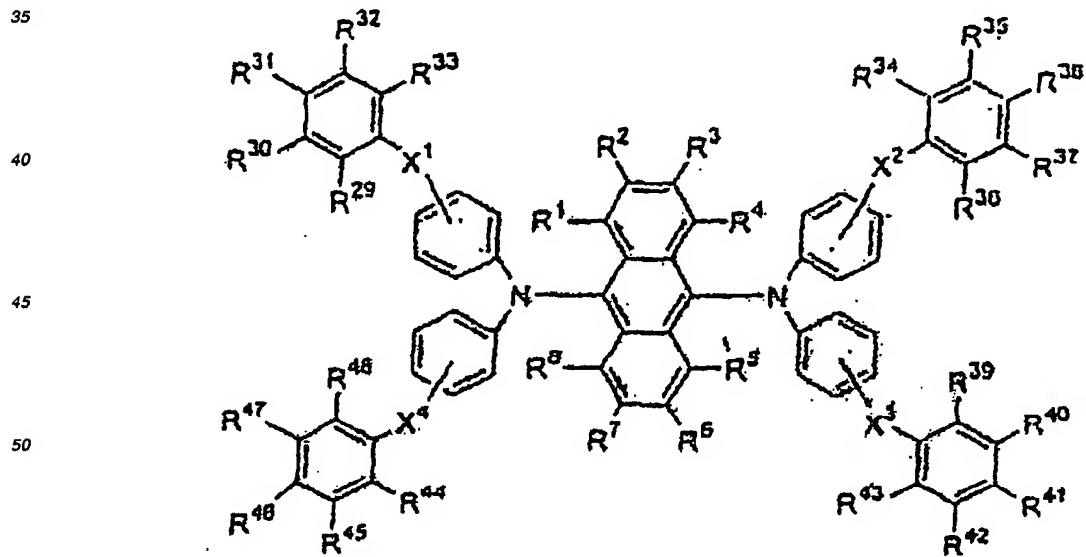


25 worin alle von R<sup>1</sup> bis R<sup>8</sup> und R<sup>29</sup> bis R<sup>48</sup> wie in Anspruch 1 definiert sind und jedes von Y<sup>1</sup> bis Y<sup>8</sup>, die gleich oder verschieden sein können, eine substituierte oder unsubstituierte Alkylgruppe mit 1 bis 20 Kohlenstoffatomen oder eine substituierte oder unsubstituierte Arylgruppe mit 6 bis 16 Kohlenstoffatomen ist.

- 30 4. Verwendung einer Verbindung nach einem der Ansprüche 1 bis 3 als ein lichtemittierendes Material in einem organischen Elektrolumineszenz-Element.

# Revendications

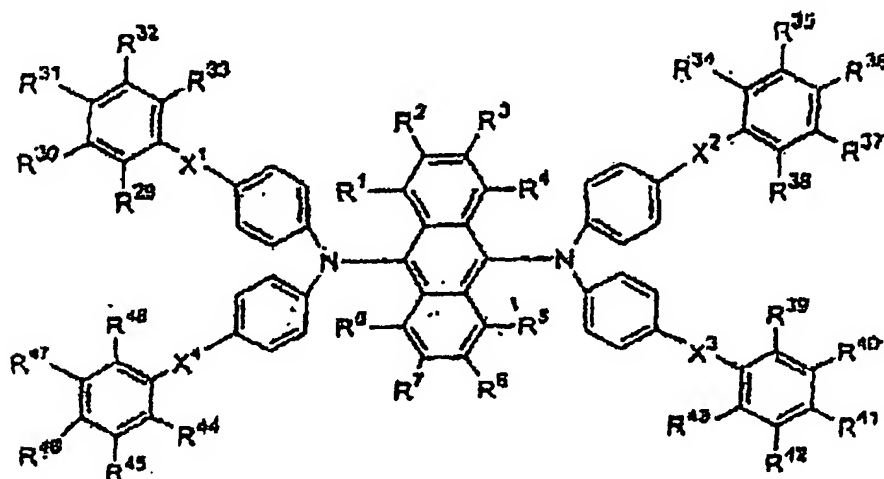
- 35 1. Composé émettant de la lumière de formule générale [3]



[ 3 ]

dans laquelle  $R^1$  à  $R^8$  et  $R^{29}$  à  $R^{48}$  sont identiques ou différents, et chacun est un atome d'hydrogène, un atome d'halogène, un groupe alkyle substitué ou non substitué, un groupe alkoxy substitué ou non substitué, un groupe aryle substitué ou non substitué ou un groupe aminé substitué ou non substitué, et  $X^1$  à  $X^4$  sont identiques ou différents et chacun est O, S, C=O, SO<sub>2</sub>, (CH<sub>2</sub>)<sub>x</sub>-O-(CH<sub>2</sub>)<sub>y</sub>, un groupe alkylène substitué ou non substitué ou un résidu alicyclique substitué ou non substitué, avec condition que x et y ne sont pas tous deux 0, et dans laquelle des substituants adjacents choisis parmi  $R^1$  à  $R^4$  et  $R^5$  à  $R^8$  forment ensemble en option un noyau aryle.

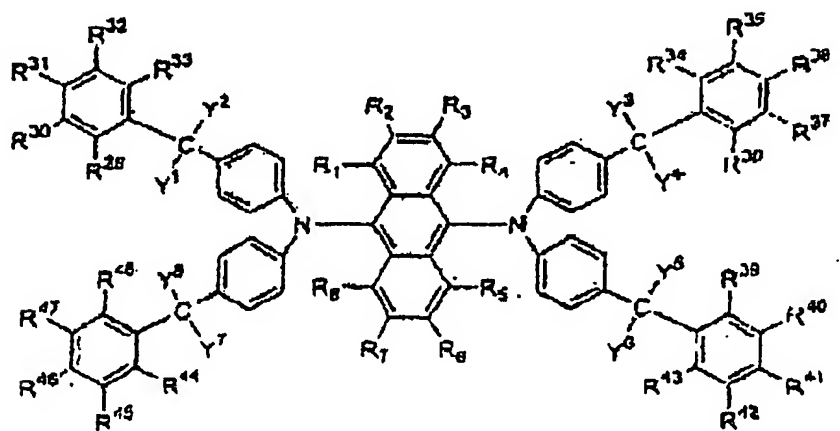
2. Composé selon la revendication 1 de formule générale [4]



[ 4 ]

dans laquelle  $R^1$  à  $R^8$ ,  $R^{29}$  à  $R^{48}$  et  $X^1$  à  $X^4$  sont tels que définis dans la revendication 1.

3. Composé émettant de la lumière de formule générale [5]



[ 5 ]

dans laquelle  $R^1$  à  $R^8$  et  $R^{29}$  à  $R^{48}$  sont chacun tels que définis dans la revendication 1 et chacun de  $Y^1$  à  $Y^8$ , qui peuvent être identiques ou différents, est un groupe alkyle substitué ou non substitué ayant 1 à 20 atomes de carbone ou un groupe aryle substitué ou non substitué ayant 6 à 16 atomes de carbone.

## EP 0 765 106 B1

4. Utilisation d'un composé selon l'une quelconque des revendications 1 à 3 comme matériau électroluminescent dans un dispositif à électroluminescence organique.

5

10

15

20

25

30

35

40

45

50

55